

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE



In re application of:

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Application No.: 10/668,964

Group Art Unit: 1711

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Examiner: Rabon Sargent

For: SOLVENT-FREE MOISTURE-CURABLE HOT MELT URETHANE RESIN  
COMPOSITION

DECLARATION UNDER 37 CFR §1.132

COMMISSIONER FOR PATENTS

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Sir:

I, Yukihiko MINAMIDA, hereby declare and state that:

1. I am a citizen of Japan, residing at Dainippon Ink and Chemicals, Inc., 35-58, Sakashita 3-chome, Itabashi-ku, Tokyo, Japan.
2. I am one of the inventors of the subject application, and I am fully familiar with the subject matter thereof as well as the references relied upon by the Examiner in the prosecution of this application.
3. I obtained a Master's degree in Polymer Science and Engineering from the Kyoto Institute of Technology in March 1988.
4. I am currently employed by Dainippon Ink and Chemicals, Inc., and began working for Dainippon Ink and Chemicals, Inc., on April 1, 1990, in research at the

urethane synthesis laboratory. Since April 2000, I have been engaged in research with the functional dispersion technology group.

5. I conducted the following experiments.

### I. Object

Tests were conducted to provide supplemental data showing that the significant effect of the present invention can be obtained only when the polyester polyol having an aromatic ring is used in combination with a morpholine ether-based crosslinking catalyst (B), sulfur atom-containing organic acid (C), and that such an effect cannot be obtained if any one of the polyester polyol having an aromatic ring, a morpholine ether-based crosslinking catalyst (B), or sulfur atom-containing organic acid (C) is lacking. It was shown that the art which does not disclose any one of the polyester polyol having an aromatic ring, a morpholine ether-based crosslinking catalyst (B), or sulfur atom-containing organic acid (C) does not suggest or teach such an effect.

### II. Synthesis

In the following examples, parts are by mass unless otherwise specified. According to the formulation shown in the following Table 1, a urethane prepolymer was synthesized by the same operation as in Example 1 of the specification. After the completion of the reaction, additives shown in the following Table 1 were added in the amount described in the same table. It is noted that data of Comparative Examples 1, Comparative Example 2, and Example 3 in Table 1 shown in page 3 is the same as those in Comparative Example 1, Comparative Example 2, and Example 2 in the original specification.

Table 1

| Composition of prepolymer<br>(Parts by mass)   | Comparison (1)        |           | Comparison (2)        |           | Comparison (3)        |           |
|--|-----------------------|-----------|-----------------------|-----------|-----------------------|-----------|
|  | Comparative Example 1 | Example 1 | Comparative Example 2 | Example 2 | Comparative Example 3 | Example 3 |
| 1,6-hexanediol adipate diol; equivalent weight of OH group=1500                            | 500                   | 500       | 500                   | 500       | 500                   | 500       |
| Neopentyl glycol terephthalate diol; equivalent weight of OH group=500                     | 250                   | 250       | 250                   | 250       | -                     | 250       |
| 1,6-hexanediol sebacate; equivalent weight of OH group=1750                                | 250                   | 250       | 250                   | 250       | 250                   | 250       |
| Isonate 143LJ (carbodiimide-modified MDI, manufactured by Mitsubishi Chemical Corporation) | 240                   | 240       | 240                   | 240       | 100                   | 240       |
| Methanesulfonic acid   | -                     | 0.625     | 0.5                   | 0.55      | 1.2                   | 1.2       |
| U-CAT2041  | 5.0                   | 5.0       | -                     | 2.2       | 9.9                   | 9.9       |
| Melt viscosity (mPa·s, 125°C)  | 14000                 | 13000     | 15000                 | 11000     | 8000                  | 10000     |
| Isocyanate group content (% by weight)   | 2.1                   | 2.2       | 2.1                   | 2.2       | 2.3                   | 2.2       |

### III. Tests

An evaluation of creep resistance at high temperature and an evaluation of thermal stability are conducted by the same method described in the specification.

### IV. Test Results

The test results are shown in the following Table.2.

Table.2

|  | Comparison (1)        |           | Comparison (2)        |           | Comparison (3)        |           |
|--|-----------------------|-----------|-----------------------|-----------|-----------------------|-----------|
|  | Comparative Example 1 | Example 1 | Comparative Example 2 | Example 2 | Comparative Example 3 | Example 3 |
| Thermal stability                                |                       |           |                       |           |                       |           |
| Melt viscosity<br>(mPa·s, 125°C)                 | 12000                 | 11000     | 13000                 | 11000     | 8000                  | 12000     |
| initial  |                       |           |                       |           |                       |           |
| after 18 hours                                   | 22000                 | 17000     | 29000                 | 16000     | 9000                  | 22000     |
| after 36 hours                                   | Gelled                | 23000     | Gelled                | 20000     | 12000                 | 35000     |
| Creep resistance at<br>60°C                      |                       |           |                       |           |                       |           |
| Immediately after<br>production of<br>specimen   | Fell off              | Fell off  | Fell off              | Fell off  | Fell off              | Fell off  |
| 1 hour after<br>standing at room<br>temperature  |                       |           |                       |           |                       |           |
|  | 20 mm                 | Fell off  | Fell off              | Fell off  | Fell off              | 9 mm      |
| 2 hours after<br>standing at room<br>temperature | Good                  | Good      | Fell off              | 9 mm      | Fell off              | Good      |
| 3 hours after<br>standing at room<br>temperature | Good                  | Good      | Fell off              | Good      | Fell off              | Good      |
| 4 hours after<br>standing at room<br>temperature | Good                  | Good      | Fell off              | Good      | Fell off              | Good      |

Regarding comparison (1):

Comparison (1) is a comparison between a composition having a urethane prepolymer with an aromatic ring, a morpholine ether-based crosslinking catalyst, and sulfonic acid (Example 1) and a composition having a urethane prepolymer with an aromatic ring, a morpholine ether-based crosslinking catalyst (Comparative Example 1).

As a result, in comparison with the thermal stability melt viscosity of the composition of Comparative Example 1, that of the composition of Example 1 declined from 12000 mPa·s to 11000 mPa·s (decline rate 8.3 %) initially, from 22000 mPa·s to

17000 mPa·s (decline rate 23 %) after 18 hours, and from the gelled state, in which the viscosity can not be measured, to 23000 mPa·s after 36 hours. Therefore, as time passes, the decline rate of the thermal stability melt viscosity of the present invention is great. Also, especially after 36 hours, it is useful that the thermal stability melt viscosity remarkably declines to a state in which the viscosity can be measured.

Also, from the result of creep resistance of the composition of Example 1, it is shown that the composition of Example 1 has sufficient function as an adhesive.

Therefore, due to the existence of sulfonic acid, the composition of the present invention has a sufficient effect of thermal stability melt viscosity and creep resistance

Regarding comparison (2):

Comparison (2) is a comparison between a composition having a urethane prepolymer with an aromatic ring, a morpholine ether-based crosslinking catalyst, and sulfonic acid (Example 2) and a composition having a urethane prepolymer with an aromatic ring and sulfonic acid (Comparative Example 2).

As a result, in comparison with the thermal stability melt viscosity of the composition of Comparative Example 2, that of the composition of Example 2 declined from 13000 mPa·s to 11000 mPa·s (decline rate 15 %) initially, from 29000 mPa·s to 16000 mPa·s (decline rate 45 %) after 18 hours, and from the gelled state, in which the viscosity can not be measured, to 20000 mPa·s after 36 hours. Therefore, as time passes, the decline rate of the thermal stability melt viscosity of the present invention is great. Also, especially after 36 hours, it is useful that the thermal stability melt viscosity remarkably declines to a state in which viscosity can be measured.

Also, from the result of creep resistance of the composition of Example 2, it is shown that the composition of Example 2 has sufficient function as an adhesive. In contrast, it is shown that the composition of Comparative Example 2 lacks sufficient function as an adhesive

Therefore, due to existence of a morpholine ether-based crosslinking catalyst, the composition of the present invention has a sufficient effect of thermal stability melt viscosity and creep resistance

Regarding comparison (3):

Comparison (3) is a comparison between a composition having a urethane prepolymer with an aromatic ring, a morpholine ether-based crosslinking catalyst, and sulfonic acid (Example 3) and a composition having a urethane prepolymer without an aromatic ring, a morpholine ether-based crosslinking catalyst, and sulfonic acid (Comparative Example 3).

As a result, the composition of Example 3 has sufficient thermal stability melt viscosity which is a little higher than those of Examples 1 and 2. Also, from the result of creep resistance of the composition of Example 3, it is shown that the composition of Example 3 has sufficient function as an adhesive.

In contrast, the thermal stability melt viscosity of the composition of Comparative Example 3 is lower than that of Example 1. However, from the result of creep resistance of the composition of Example 3, it is shown that the composition of Comparative Example 3 lacks sufficient function as an adhesive.

Therefore, due to the existence of an aromatic ring in a urethane prepolymer, the composition of the present invention has a sufficient effect of thermal stability melt viscosity and creep resistance.

## V. Conclusion

As indicated above, the solvent-free moisture-curable hot melt urethane resin composition, in which the polyester polyol having aromatic ring is used in combination with a morpholine ether-based crosslinking catalyst (B), sulfur atom-containing organic acid (C), has significant effects as claimed in claim 1 of the present application.

We believe that the art which does not disclose any one of the polyester polyol having aromatic ring, a morpholine ether-based crosslinking catalyst (B), or sulfur atom-containing organic acid (C) does not suggest or teach such an effect.

6. I understand fully the content of this declaration.
7. The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed

to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

8. Further declarant saith not.

Yukihiko Minamida  
Yukihiko MINAMIDA

2006, 2 . 24  
Date